

Supporting Information

A Hairpin-Contained I-Motif Guided DNA Nanoantenna for Sensitive and Specific Sensing of Tumor Extracellular pH

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Contents

1. Oligonucleotides used in the work. (Table S1)
2. Secondary structure of I-strand, pH-Apt-YNA, and pH-YNA, respectively. (Fig. S1)
3. Validation of pH-YNA probe by 2% agarose gel electrophoresis. (Fig. S2)
4. Verification the formation of i-motif structure in different probes at pH 5.6 and 7.4 by UV absorption spectra. (Fig. S3)
5. Verification the formation of i-motif structure in different probes at pH 6.2 and 7.4 by fluorescence spectra. (Fig. S4)
6. UV response of pH-Apt-YNA at different pH. (Fig. S5)
7. Co-localization analysis of pH-Apt-YNA and cell membrane dye. (Fig. S6)

Table S1. Oligonucleotides used in this work.^a

Name	Sequence (5' to 3')
I-strand	BHQ ₂ -CCC CCT CCC CCA TAT TAA ACT CTA ATA ACC CCC TCC CCC
I-strand-G	BHQ ₂ -CCC CCT CCC CCA TAT TAA ACT CTA ATA ACC CCC TCC CCC- Rhodamine Green
Apt-Y-R	ACT TTG GGT AGG GTG AGG TGG TT AAA AGA <u>ACG GCG CAT GAC CGA</u> <u>AT</u> TTT <i>GGT GGT GGT GGT TGT GGT GGT GGT GG</i>
Y-R	ACT TTG GGT AGG GTG AGG TGG TT AAA AGA <u>ACG GCG CAT GAC CGA</u> <u>AT</u>
Y-L-G	<u>ATT CGG TCA TGC GCG AT</u> TTT AAA AT GGT GGA GTG GGG TGG AGG CTAC-Rhodamine Green
Y-L	<u>ATT CGG TCA TGC GCG AT</u> TTT AAA AT GGT GGA GTG GGG TGG AGG CTAC

^a The red letters in I-strand represent hairpin sequences. The underlined letters represent the complementary base sequences. The italics in Apt-Y-R are aptamer fragments of AS1411 for MCF-7 cells.

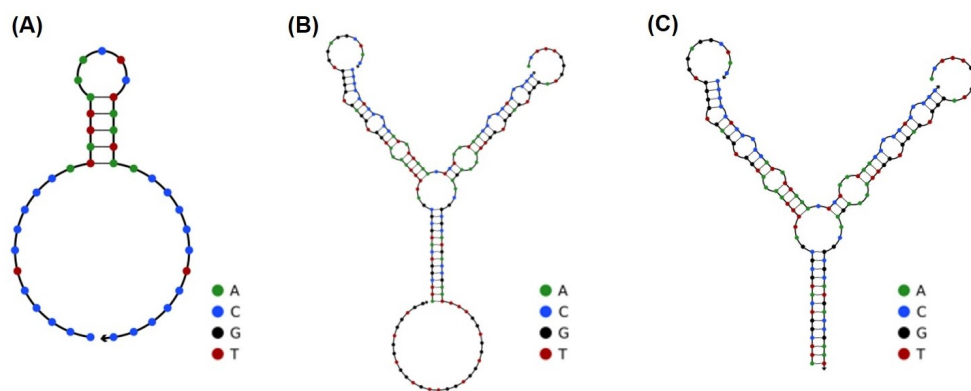


Fig. S1 The secondary structure of (A) I-strand, (B) pH-Apt-YNA, and (C) pH-YNA simulated by NUPACK (<http://www.nupack.org/>), respectively. The concentration of Na^+ and Mg^{2+} was 130 mM and 5 mM, respectively. And the simulated temperature was set at 37 °C.

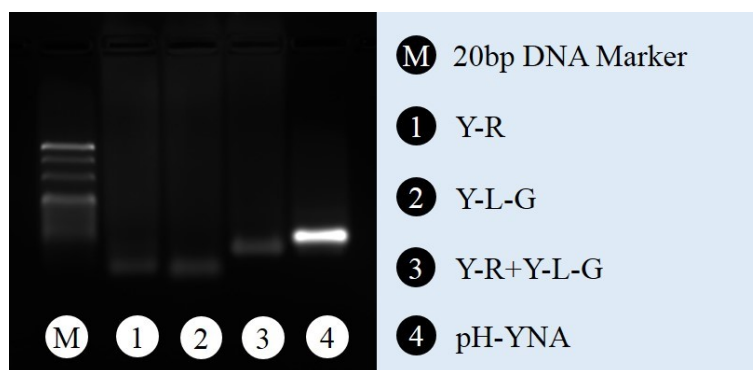


Fig. S2 Validation of pH-YNA (pH 7.4) by 2% agarose gel electrophoresis. The probe concentration was 1 μ M.

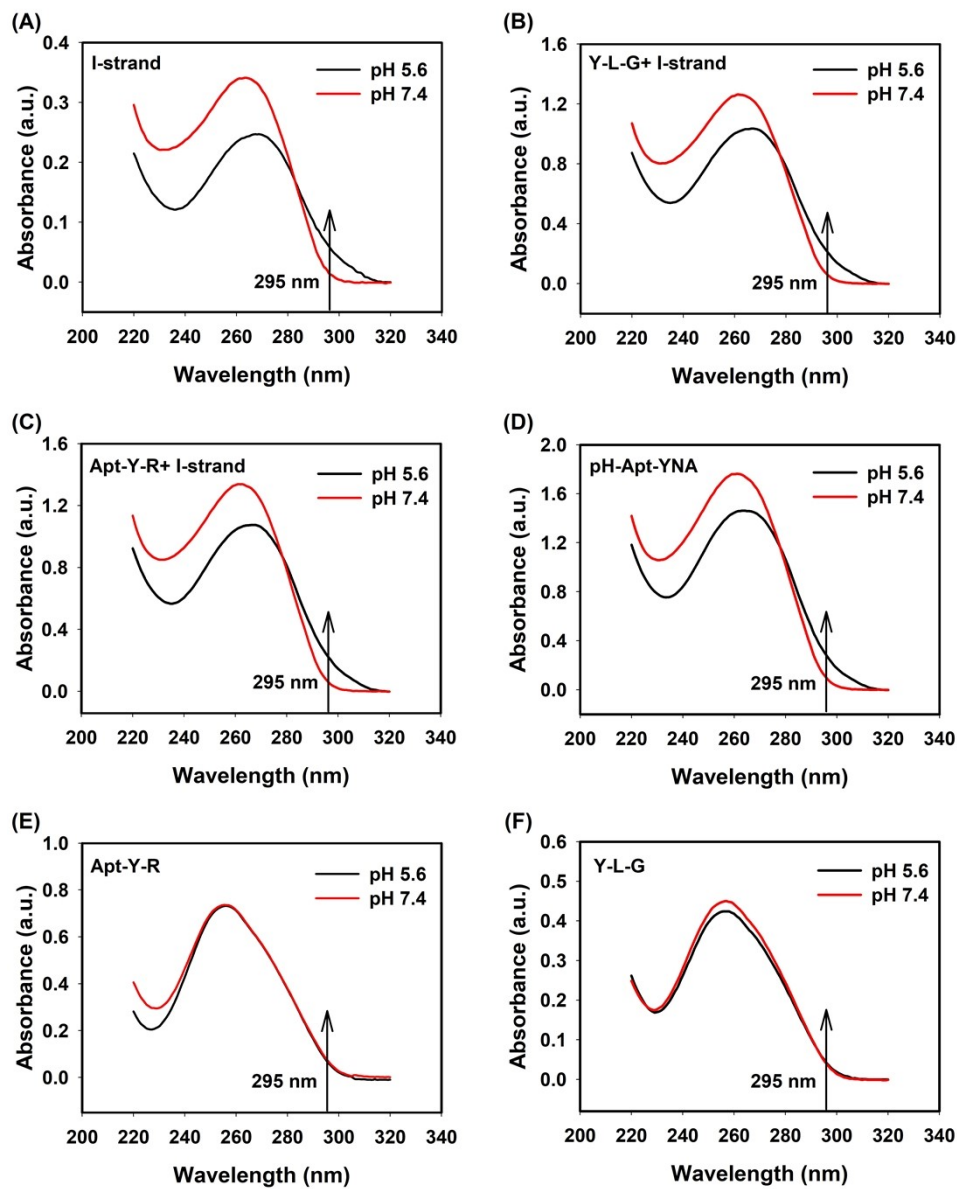


Fig. S3 Verification the formation of i-motif structure in different probes (500 nM) under various pH conditions (pH 5.6 and 7.4) by UV absorption spectra. (A) I-strand, (B) Y-L-G+I-strand, (C) Apt-Y-R+I-strand, (D) pH-Apt-YNA (Apt-Y-R+Y-L-G+I-strand), (E) Apt-Y-R, and (F) Y-L-G, respectively.

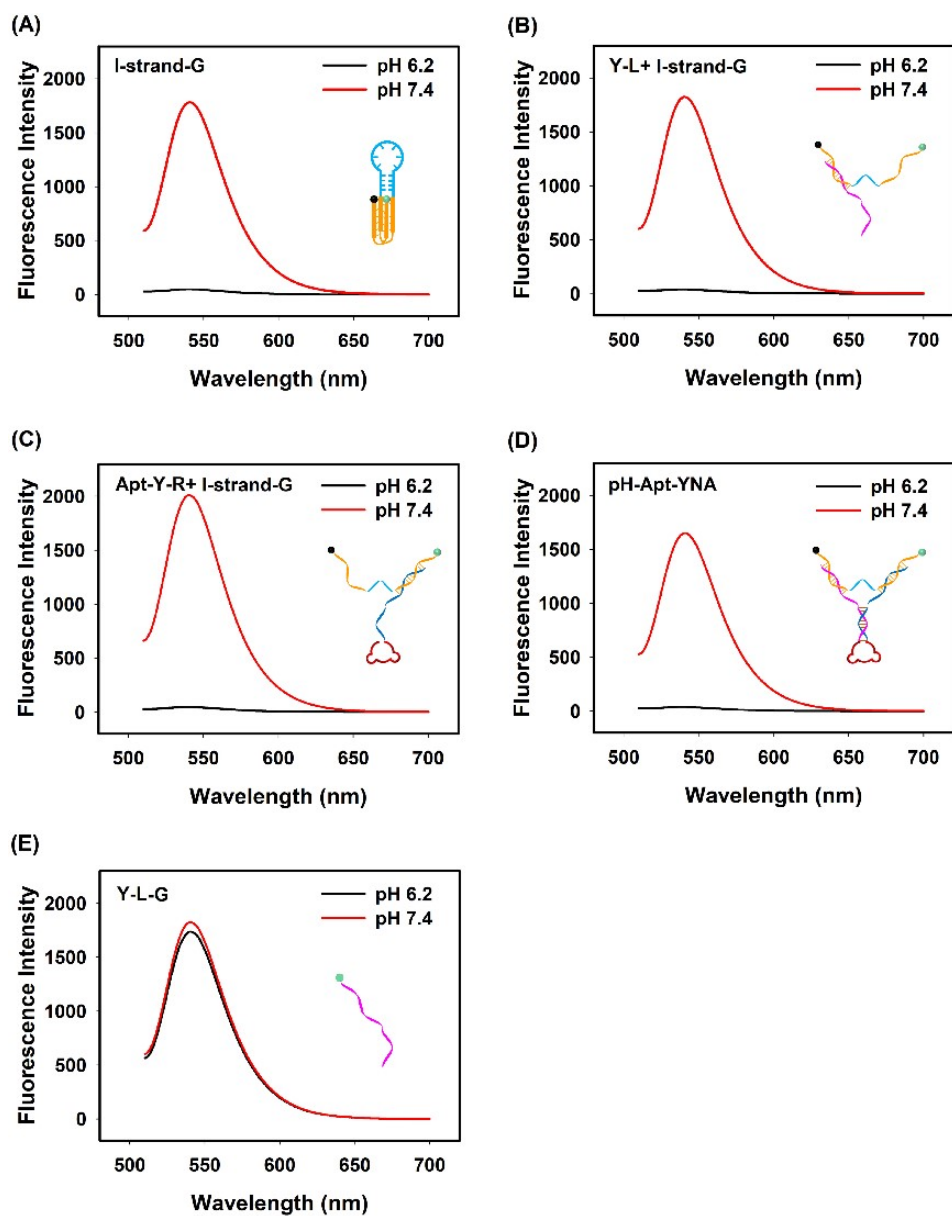


Fig. S4 Verification the formation of i-motif structure in different probes (100 nM) under various pH conditions (pH 6.2 and 7.4) by fluorescence spectra. Ex = 498 nm, Em = 510-700 nm. (A) I-strand-G, (B) Y-L+I-strand-G, (C) Apt-Y-R+I-strand-G, and (D) pH-Apt-YNA (Apt-Y-R+Y-L+I-strand-G), (E) Y-L-G, respectively.

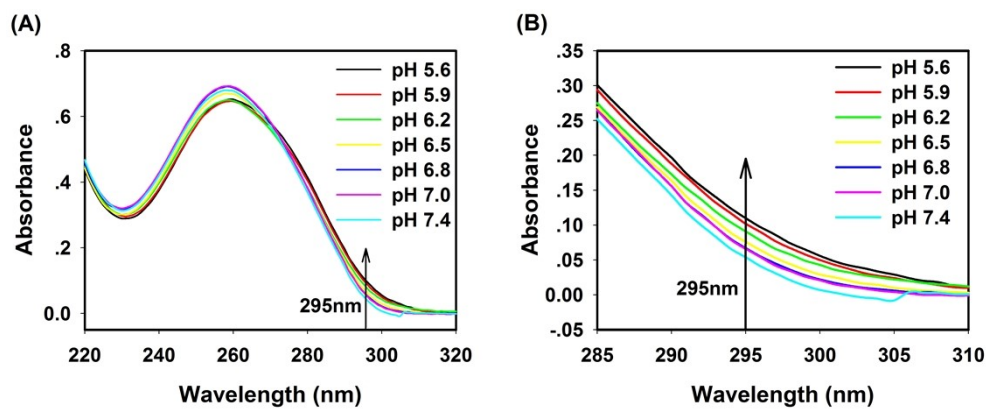


Fig. S5 (A) Verification the formation of i-motif structure in 200 nM pH-Apt-YNA under different pH conditions (pH 5.6, 5.9, 6.2, 6.5, 6.8, 7.0 and 7.4) by UV absorption spectra. The (B) shows enlarged image of Figure (A) at the wavelength of 285-310 nm.

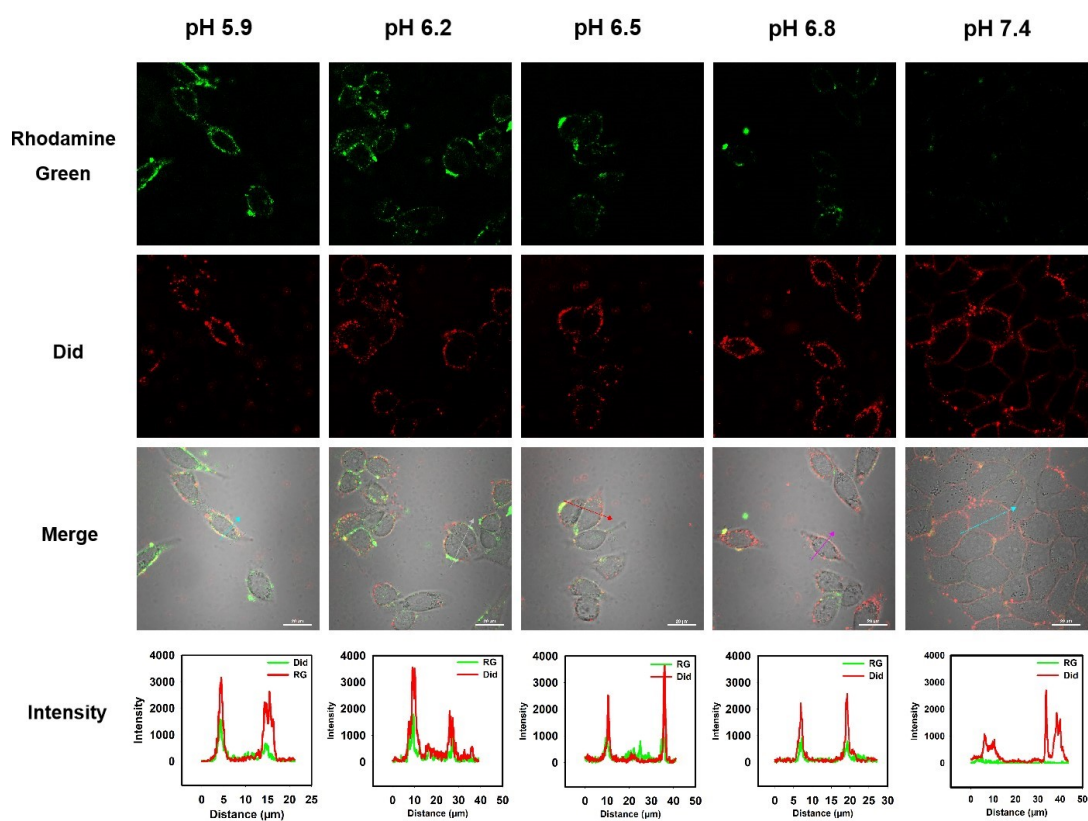


Fig. S6 Fluorescence imaging of MCF-7 cells treated with pH-Apt-YNA under different pH values (pH 5.9, 6.2, 6.5, 6.8, and 7.4) and co-localization analysis of pH-Apt-YNA and cell membrane dye (Did). Excitation and emission wavelengths were described as follows. Rhodamine Green (RG): Ex = 488 nm, Em = 505-560 nm bandpass; Cy5: Ex = 640 nm, Em = 663-738 nm bandpass, bar=20 μm .